

# Multiplexed holograms recorded in a low toxicity Biophotopol photopolymer

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## ABSTRACT

Multiplexed diffraction gratings were recorded in 300  $\mu\text{m}$  thick layers of Biophotopol photopolymer by using peristrophic multiplexing schema. Thirteen sinusoidal phase gratings were stored in a low toxicity recording medium. The diffraction efficiency conservation of the multiplexed diffraction efficiency obtained was studied along the time.

**Keywords:** Multiplex holography, Biomaterials, Optical storage-recording materials, Stability

## 1. INTRODUCTION

Photopolymers are useful for different holographic applications such as memories, optical elements or sensors. These materials are light sensitive and have demonstrated its potential use as a holographic recording materials due to the advantages of chemical versatility, easy processability and low cost offered by organic materials. These photopolymers have good properties, in particular: good energetic sensitivity compared with other available materials; the possibility of easily adapting their spectral sensitivity to the type of laser used by simple changing the sensitizer dye; high diffraction efficiency, together with an acceptable resolution and signal/noise ratio. The development of suitable recording material has been one of the main goals in the development of holographic data storage. There are many types of photopolymers that may be differentiated by the type of binder, since this component determines to a great extent the choice of monomer, dye and initiator used in the photopolymer, most of them have some drawbacks that prevent their use in practical holographic systems.

Traditionally, polyvinyl alcohol/acrylamide has been considered as a versatile holographic recording material; however, this type of photopolymer has low environmental compatibility. Instead, our team has been developing during the last years a biodegradable and biocompatible photopolymer called Biophotopol which has been showed high diffraction efficiency (with reported values near 90%) in 300  $\mu\text{m}$  thick layers. Recently, the Biophotopol photopolymer capability as a data recording material has been demonstrated using two different peristrophic multiplexing techniques<sup>1</sup>. These techniques are commonly used in holographic data storage experiments<sup>2-4</sup>. Peristrophic multiplexing is a very useful tool especially when the holographic recording medium is relatively thin. It is usually implemented by combining of two or more multiplexing techniques that increase the storage density. There is no loss of the previous grating when a new grating is recorded at the same point.

In this work, thirteen gratings have been recorded by using the parallel multiplexing technique at a single location. The rotation axis is parallel to the sample plane and thus the angle of incidence of the reference and object beams vary continuously during storage of the holograms. Each hologram is stored at an angle of incidence slightly different to that of the previously recorded one.

## 2. EXPERIMENTAL

### 2.1 Preparation of the material

Photopolymer layers consist of a binder with one or more monomers, an electron donor, and a dye sensitizer. The Biophotopol composition was based on previous studies where sodium acrylate (NaAO) was used as the monomer.

Table 1 shows the component concentrations of the Biophotopol photopolymer to obtain 300  $\mu\text{m}$  thick layers. The composition consist of water as solvent, triethanolamine (TEA) as coinitiator and plasticizer, riboflavin 5'-monophosphate sodium salt (RF) as dye and polyvinyl alcohol (PVA) as binder ( $M_w = 130000$  g/mol, degree of hydrolysis = 87.7%) with the corresponding monomer (NaAO). The quantity of solution used was calculated from previous studies in order to obtain 300  $\mu\text{m}$  thick Biophotopol photopolymer film giving maximum diffraction efficiency<sup>5</sup>.

Table 1. Molarity composition of the Biophotopol photopolymer. PVA in percentage

NaAO (M)	PVA (w/v)	TEA (M)	RF (M)
0.34	13.4	$9.2 \times 10^{-3}$	$1.0 \times 10^{-3}$

After recording the gratings not all the dye in the photopolymer is consumed and the reaction continues if the hologram is exposed to incoherent light. One way to avoid this reaction is to remove the excess dye after recording the grating. Therefore, after the initial reconstruction ( $t = 0$  h), we exposed the sample to a 13.5 W (875 lumen at 6500K) LED lamp for 20 minutes. This curing process does not erase the grating because the dye that remains in the exposed zones is negligible. This process only affects the remaining dye in the unexposed zones and therefore the grating renders more stable over time<sup>6</sup>. The cured gratings were stored in ambient conditions where the temperature and relative humidity change depending on the season.

### 2.2 Holographic setup

Multiplexed gratings were recorded at a constant exposure intensity of 3 mW/cm<sup>2</sup> provided by an Argon laser (BeamLok 2060 by Spectra-Physics) tuned at a wavelength of 488 nm with continuous laser exposure. The polarized beam was split into two secondary beams with an intensity ratio of 1:1 and a diameter of 1.5 cm. The angle of incidence of both the object and the reference beam at the recording medium were 17.1° (measured in air). This angle was used to obtain a spatial frequency of 1200 lines/mm.

For peristrophic multiplexing studies, the sample was mounted on a motorized rotation stage. Holograms were read out with an angular resolution of 0.02°. All components (shutter, rotation stages and detectors) were connected to a computer and controlled by a LabVIEW program.

Gratings were peristrophic multiplexed by rotating the parallel axis. The system allows multiple gratings to be recorded in the same volume of material. Each grating is recorded at an angle that is Bragg mismatched from its neighboring gratings and reconstructed away from the detector, permitting a new grating to be recorded. In order to prevent grating overlapping during the storage process the angle between consecutive gratings selected was 3°. This ensures there is sufficient separation between the gratings to enable them to be subsequently reconstructed independently.

The exposure time was controlled by placing an electronic shutter in front of the Ar laser. No pre-exposure energy was used to reduce the inhibition period because the polymerization process began almost instantaneously, in both compounds. The analysis in real time of grating formation and the angular multiplexed gratings reconstruction processes were made using a HeNe laser. Diffraction efficiency (DE) and transmission efficiency (TE) were calculated as the intensity of the diffracted and transmitted beams divided by the incident beam, respectively.

## 3. RESULTS AND DISCUSSION

### 3.1 Multiplexing

Thirteen volume transmission holograms were stored at a single location using the peristrophic multiplexing technique with the rotation axis parallel to the sample plane as can be seen in Fig. 1. The first grating stored was at zero degrees

(unslanted grating). The following gratings were stored at different angles from zero (slanted gratings). From the Fig 1 it can be seen that the slanted gratings recorded have a shift from their original register angle. This displacement is a consequence of the shrinkage that appears due to a film thickness variation<sup>2,7</sup>. In some gratings, it is also possible to see the secondary lobes that appear in the slanted gratings ( i.e. for grating registered at  $-6^\circ$  in the left part of the principal lobe and for grating registered at  $3^\circ$  in the right part of the lobe).

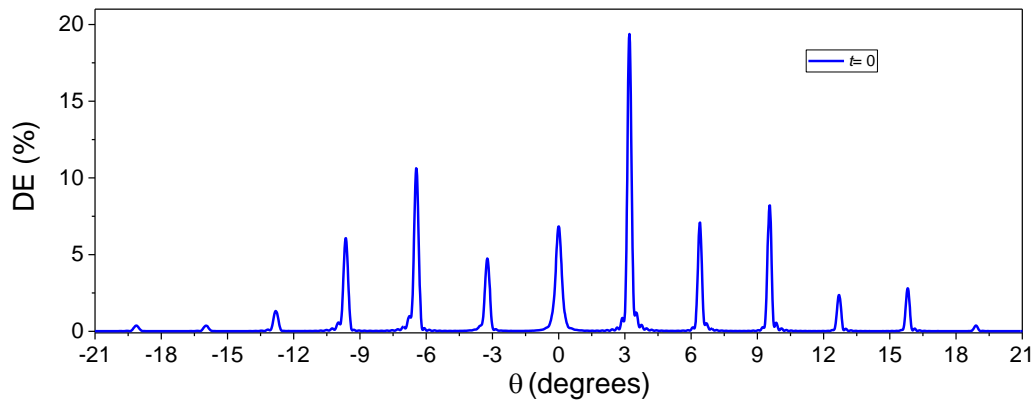


Fig. 1. Grating diffraction efficiency (DE) versus the axis parallel to the sample ( $\theta$ ).

The maximum diffraction efficiencies ( $DE_m$ ) obtained for the thirteen gratings are not uniform because a schedule time procedure was not used.  $DE_m$  values over 5% are obtained only for the first 7 gratings recorded. The last 6 gratings have lower  $DE_m$  values.

### 3.2 Grating stability

In order to determine the storage life behaviour of the gratings recorded in the Biophotopol photopolymer the DE reconstruction was performed over the time. For this purpose the previous thirteen multiplexed gratings recorded have been assessed.

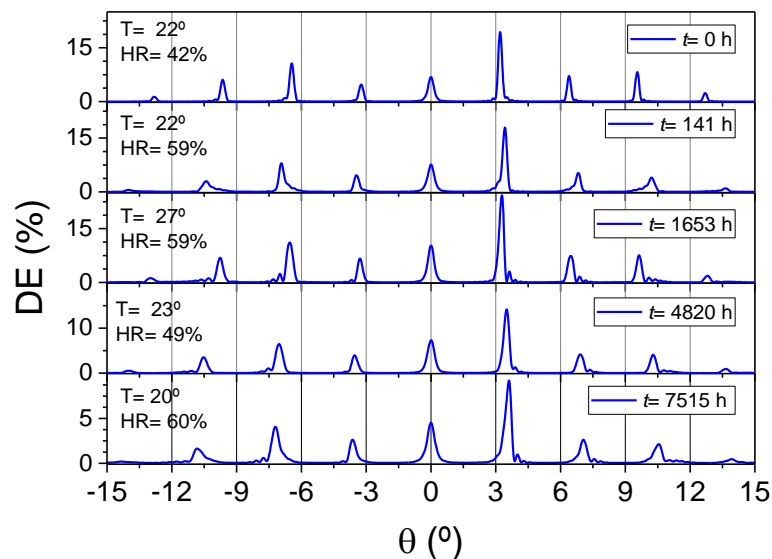


Fig. 2. Angular reconstruction of multiplexed gratings along the parallel axis ( $\theta$ ) at different times ( $t$ ).

Only 7 of the 13 gratings have been analyzed due to the poor  $DE_m$  obtained in the gratings recorded in the extremes. Over a period of ten months the  $DE_m$  values have fluctuated around the initial values. Fig. 2 shows the gratings reconstructed for five representative time measurements. The temperature ( $T$ ) and relative humidity ( $HR$ ) of the storage place for each reconstruction are included. It is possible to observe the changes in the  $DE$  of the analyzed gratings and also in the shapes of the reconstruction curves.

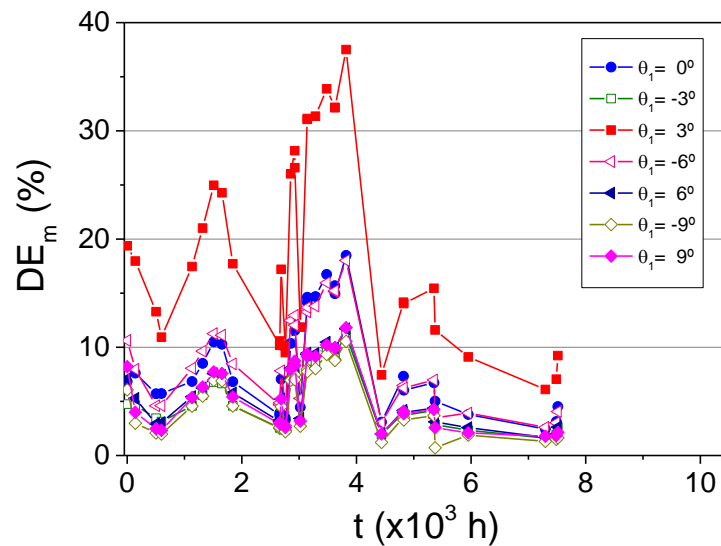


Fig. 3. Maximum diffraction efficiency ( $DE_m$ ) versus reconstruction time ( $t$ ) for each grating.

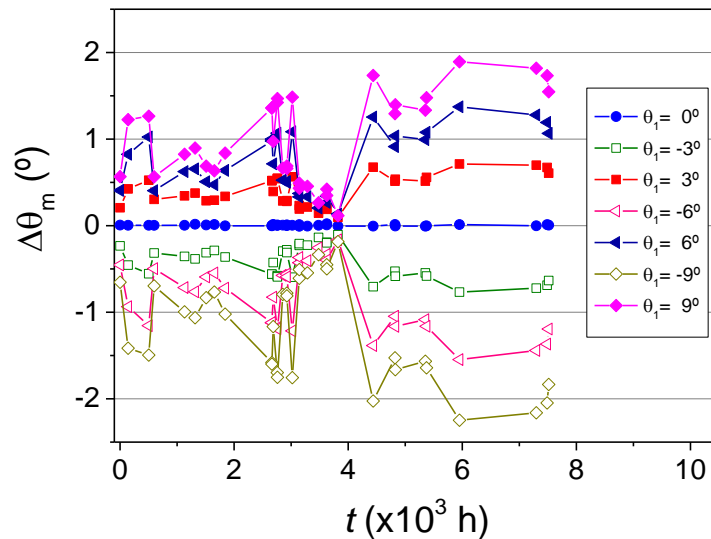


Fig. 4. Angle shift of the maximum  $DE$  versus reconstruction time ( $t$ ) for each grating.

Fig. 3 shows the  $DE_m$  evolution of the gratings versus time accurately. All the curves on this graph have the same shape, which indicates that the  $DE$  of the gratings evolves in a similar manner. As we can see, the  $DE_m$  changes over time and this variation depends on the initial  $DE_m$  value. The higher the initial  $DE_m$  value the greater the variation over time

produce. The greater variation was obtained for  $\theta = 3^\circ$  geometry, which had an initial  $DE_m$  higher than the other gratings. For this geometry, the  $DE_m$  arrived to around 38% ( $t = 3820$  h) from 19% ( $t = 0$  h).

Fig. 4 shows the angular displacement ( $\Delta\theta_m$ ) of  $DE_m$  versus time. This displacement is related to shrinkage<sup>8</sup> as we assessed and quantified in previous studies<sup>7</sup>. Firstly,  $\Delta\theta_m$  is close to zero for the unslanted grating stored at the zero position ( $\theta = 0^\circ$ ). The slanted gratings —stored to the right and left of the zero position— present a variation in  $\Delta\theta_m$  and this variation is symmetrical relative to the zero position. Comparing Fig. 3 and Fig. 4  $\Delta\theta_m$  increases as  $DE_m$  decreases and vice versa. Therefore, this variation depends on the initial  $DE_m$ , temperature (T) and relative humidity (HR) conditions. It also can be seen that for  $t = 6 \cdot 10^3$  h the angular position of the DE values of each grating reached his maximum  $\Delta\theta_m$  value. That corresponds to the minimum  $DE_m$  obtained for each grating, as it can be seen in Fig. 3.

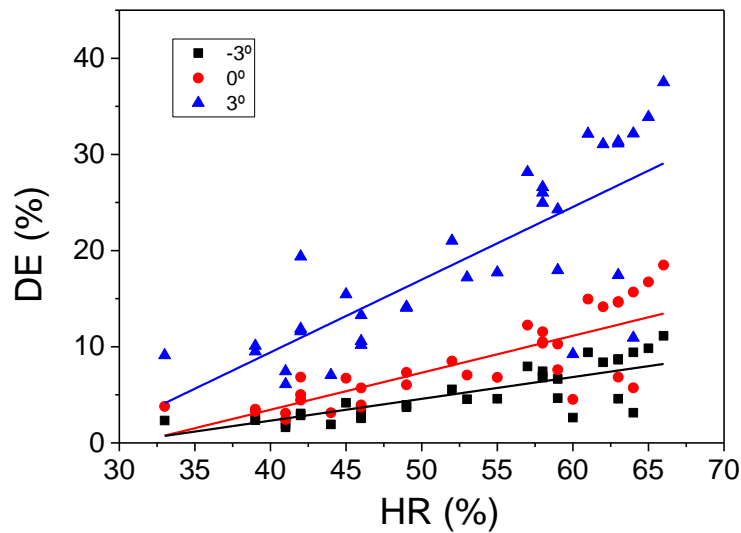


Fig. 5. Maximum diffraction efficiency ( $DE_m$ ) versus relative humidity (HR) for (-3, 0, and 3 geometries). A tendency line is added for each grating in order to show the trending of the  $DE_m$  values.

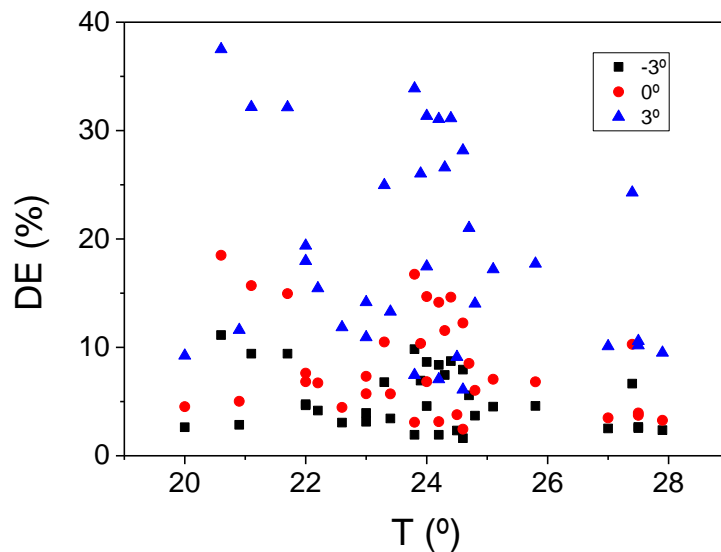


Fig. 6. Maximum diffraction efficiency ( $DE_m$ ) versus temperature (T) for (-3, 0, and 3 geometries).

Fig. 5 shows the  $DE_m$  versus relative humidity. Only  $DE_m$  of three gratings have been represented. In all cases the  $DE_m$  increase when the HR increase, thus it can be seen that the DE is correlated with the HR. For example, for  $t= 3820$  h the laboratory achieved the highest relative humidity (HR = 65%) the values DE obtained were the highest (i.e.  $DE_m= 38\%$  for  $\theta= 3^\circ$  geometry). A guide-line has been added to see the linear tendency.

The last reconstruction measurement ( $t= 7515$  h) shows a small rise in the DE. This increase is due to the fact that the sample was introduced in a climate chamber for one hour at a controlled temperature and relative humidity ( $T=20^\circ$  and  $HR=60\%$ ). With this process we wanted to prove that the gratings had not deteriorated due to the low relative humidity given in the laboratory during the last three months. The low  $DE_m$  ( $t= 7515$  h,  $HR= 60\%$ ) obtained compared with other  $DE_m$  values at similar HR (i.e.  $t= 1653$  h,  $HR= 59\%$ ) conditions could be due to the short time that the sample was exposed to the HR value of 60%. This can be seen in the greater dispersion of the  $DE_m$  when the HR is high. In future studies we will analyze the DE dependence with the HR controlled.

Fig. 6 shows the  $DE_m$  versus temperature. As it can be seen the  $DE_m$  does not show a dependency with the temperature.

Therefore, it was possible to restore or even increase the initial  $DE_m$  by changing the value of HR. These variations could allow us to use Biophotopol photopolymer as a sensor. Other authors has recently demonstrated similar dependency for diacetone acrylamide-based gratings in the range of 20-60% HR<sup>9</sup>.

## 4. CONCLUSIONS

In this study we demonstrated the potential use of the Biophotopol photopolymer as a multiplexing material. Moreover, it is also analyzed the variability of the holographic behaviour with the relative humidity and temperature. The humidity dependence is greater than the temperature dependence. The DE variability depends on the initial DE value and at the same time the variability of the DE is related to the shift of the angular position in the slanted gratings. Its performance is comparable to other photopolymers, with the added advantage of its low toxicity. A possible future study consists in using a crosslinker or sealing the samples to enhance the stability of the gratings.

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